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(71)Applicant : IBIDEN CO LTD

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(54) PRODUCTION OF CATALYST CARRIER

(57)Abstract:

PURPOSE: To produce a catalyst carrier made of a porous silicon carbide sintered body having a high strength, a large specific surface area and a large pore volume.

CONSTITUTION: Silicon carbide powder having 0.1-5m²/g specific surface area and contg. 1.0-5% impurity components preferably including 0.1-0.5% (expressed in terms of boron) boron compd. is used as starting material, compacted into a desired shape, dried and fired in the temp. range of 1,600-2,200° C to produce the objective catalyst carrier.

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(54)【発明の名称】 触媒担体の製造方法

(57)【要約】

【目的】本発明は高い強度、大きな比表面積と細孔容積を有する多孔質炭化ケイ素焼結体からなる触媒担体の製造方法を提供することを目的とする。

【構成】本発明は、比表面積が $0.1 \sim 5 \text{ m}^2/\text{g}$ 、不純物成分が $1.0 \sim 5\%$ の炭化ケイ素粉末、さらに好ましくは不純物成分中にホウ素化合物が金属換算で $0.1 \sim 0.5\%$ 含有している炭化ケイ素粉末を出発原料とし、これを所望の形状に成形、乾燥後、 $1600^\circ\text{C} \sim 2200^\circ\text{C}$ の温度範囲で焼成することを特徴とする触媒担体の製造方法である。

【特許請求の範囲】

【請求項1】 比表面積が $0.1 \sim 5 \text{ m}^2/\text{g}$ 、不純物成分が $1.0 \sim 5\%$ の炭化ケイ素粉末を出発原料とし、これを所望の形状に成形、乾燥後、 $1600^\circ\text{C} \sim 2200^\circ\text{C}$ の温度範囲で焼成することを特徴とする触媒担体の製造方法。

【請求項2】 不純物成分中にホウ素化合物が金属換算で $0.1 \sim 0.5\%$ 含有していることを特徴とする請求項第1項記載の触媒担体の製造方法。

【発明の詳細な説明】

【0001】

【産業上の利用分野】本発明は高い強度、大きな比表面積と細孔容積を有する多孔質炭化ケイ素焼結体からなる触媒担体の製造方法に関する。

【0002】

【従来の技術】炭化ケイ素は高い硬度、優れた耐摩耗性、優れた耐酸化性、優れた耐蝕性、良好な熱伝導率、低い熱膨張率、高い耐熱衝撃性、並びに、高温での高い強度等の化学的および物理的に優れた特性を有し、メカニカルシールや軸受け等の耐摩耗材料、高温炉用の耐火材や熱交換器または燃焼管等の耐熱構造材料、酸およびアルカリ等の強い腐蝕性を有する溶液のポンプ部品等の耐蝕性材料として広く使用可能な材料である。

【0003】一方、これらの性質を有する炭化ケイ素と、その結晶が形成する通気性を有するところの気孔、すなわち開放気孔とからなる多孔質炭化ケイ素質焼結体は、前記炭化ケイ素の特徴を生かして、高温雰囲気、酸化性雰囲気及び/または腐蝕性雰囲気下における耐熱、耐蝕性物質分離材料として利用可能であり、例えば内燃機関の排気ガス、特にディーゼルエンジンの排気ガス等の高温気体中に含まれる微粒子カーボン等の微粒子物質の除去のために使用されるフィルターとして利用しうる。更に、この多孔質炭化ケイ素フィルター表面に、酸化反応用触媒成分を担持せしめた場合には、可燃性のカーボン微粒子を燃焼せしめ、ガスに転化させることも可能で、この場合、この多孔質炭化ケイ素フィルターは、耐熱・耐蝕触媒としても機能することになる。

【0004】また最近、主として環境汚染防止の観点から、内燃機関やガスタービン用ボイラーなどの工業用燃焼装置の分野において、低 NO_x 燃焼技術の研究開発が行われており、その一つとして燃焼触媒を用いる触媒燃焼技術が注目を集めている。この触媒燃焼技術の開発における最も重要な要素は、触媒の開発であり、触媒の開発においては活性な酸化反応用触媒物質の開発と並んで、活性成分を分散、担持するための担体の開発が極めて重要である。

【0005】燃焼触媒用担体においては、触媒表面で進行する燃焼反応、すなわち、酸化反応を迅速に生起せしめるために表面積が大きいことに加えて、発生する反応熱を有効に伝達・除去できるような良好な熱伝導度を有

すること、および、触媒細孔内の物質移動を有効に行わせるためにガス等の流体の通過抵抗が小さいこと、すなわち、細孔容積が大きいこと、更に、成形体相互のぶつかり合いによるアブレイジョン、即ち、磨滅に強いことおよび成形体自身が十分な機械的強度を有すること、そして、これらの特性が長時間の使用に対して安定していることなど、多くの要求を満足することが必要である。このような要件は、燃焼触媒のみならず、一般に反応熱の発生を伴う化学反应用の触媒担体、あるいは高温で使用される触媒用の担体についても共通的にいえることである。

【0006】一方、多孔質炭化ケイ素質焼結体の製造方法としては、(1)骨材となる炭化ケイ素粒子にガラス質フラックス、あるいは粘土質などの結合材を加え成形した後、その成形体を前記結合材が溶融する温度で焼き固めて製造する方法、(2)粗大粒の炭化ケイ素粒子と微細な炭化ケイ素粒子を混合し成形した後、 2000°C 以上の高温で焼成して製造する方法、あるいは(3)特開昭48-39515号の発明で開示されている炭化ケイ素粉に炭素粉を加え、または加えずに炭素質バインダーを加えると共に、炭素粉および焼成時に生成されるバインダーからの遊離炭素と反応する理論量のケイ素質粉を添加して形成し、しかる後、この成形体中の炭素粉中で $1900 \sim 2400^\circ\text{C}$ に加熱して成形体中の炭素分をケイ素化することを特徴とする均質多孔性再結晶炭化ケイ素体の製造方法、(4)比表面積が $3 \text{ m}^2/\text{g}$ 以上で、ホウ素、アルミニウムおよび鉄の含有量の合計が元素に換算して 0.3 重量%以下である炭化ケイ素粉末を所望の形状に成形後、 $1600 \sim 2200^\circ\text{C}$ で焼成して製造する方法等が従来知られている。

【0007】

【発明が解決しようとする課題】しかしながら、前述した従来技術には次の問題がある。

(1)骨材となる炭化ケイ素粒子にガラス質フラックスあるいは粘土質などの結合材を加え成形した後、その成形体を前記結合材が溶融する温度で焼き固めて製造する方法は、結合材が低温($1000 \sim 1400^\circ\text{C}$ 前後)で溶融するため、多孔質体はこの温度域、特にガラス転移温度付近で変化する著しく強度が低下するだけでなく、耐薬品性、耐酸化性が要求される分野における使用が限られるという欠点がある。

【0008】(2)粗大粒の炭化ケイ素粒子と微細な炭化ケイ素粒子を混合し成形した後 2000°C 以上の高温で焼成して製造する方法、あるいは、(3)特開昭48-39515号の発明で開示されている炭化ケイ素粉に炭素粉を加え、または加えずに炭素質バインダーを加えると共に、この炭素粉及び焼成時に生成されるバインダーからの遊離炭素と反応する理論量のケイ素質粉を添加して形成し、しかる後、この成形体中の炭素粉中で $1900 \sim 2400^\circ\text{C}$ に加熱して成形体中の炭素分をケイ素

化することを特徴とする均質多孔性再結晶炭化ケイ素体の製造方法は、多孔質炭化ケイ素骨材とその骨材を被覆して、骨材同志を結合する炭化ケイ素質結合材あるいは炭素質結合材および間隙とが構成される。前記多孔質の間隙、すなわち開放気孔は殆ど成形時に骨材粒子となる粗大粒子を多く必要としその結果骨材粒子の接点数が少なくなり、多孔質の強度は著しく低下し、しかも比表面積は $0.5 \text{ m}^2/\text{g}$ 以下で著しく小さいものとなる。

【0009】一方、強度の高い多孔質体とするためには骨材の粒度配合を粗粒と中程度／または微粒子と適度に混合し形成することが必要でありその結果、多孔質体の細孔容積は高々 0.1 ml/g で著しく小さく極端な場合、一部の開放気孔が閉塞してしまう傾向がある。このため、このような、多孔質体を流体が通過する際の抵抗は著しく高くなり、物質分離用フィルターや、触媒担体素として利用する場合、著しく不利益となる。従って、触媒担体として好適な特性を有する炭化ケイ素質焼結体即ち、取扱いが容易な強度を有し、しかも細孔容積が $0.2 \text{ m}^2/\text{g}$ より大きく、比表面積が $3 \text{ m}^2/\text{g}$ より大きな多孔質炭化ケイ素質焼結体は存在しない。

【0010】(4) 前記のような性質を有する多孔質体を製造する方法として出願人は先に提案した方法があるが、この方法は、比表面積が $3 \text{ m}^2/\text{g}$ 以上で、ホウ素、アルミニウム、および鉄の含有量が合計で 0.3 重量%以下のものを用い、 $1400 \sim 2000^\circ\text{C}$ で焼成する方法は高純度で、且つ超微粉炭化ケイ素を出発原料としているために著しくコストアップとなるだけでなく、成形体の乾燥収縮率が大きくなって、乾燥亀裂が発生し易い欠点をする：ディーゼル・エンジン排ガス中のパーティキュレート・フィルターのよう気孔径が $15 \mu\text{m}$ 前後が適切であるのに対して、数 μm 前後又はそれ以下の気孔径のものしか得られない。気孔径を大きくする方法としては焼成温度を 2000°C 以上で行えば良いが、前記方法では異常粒成長等により強度劣化を起すこととなる等の欠点を有していた。

【0011】

【課題を解決するための手段】本発明は、前記従来の欠点を解消し、かつ改善して、耐熱触媒担体として必要な特性を有する多孔質炭化ケイ素質焼結体からなる触媒担体を供給するために、比表面積が $0.1 \sim 5 \text{ m}^2/\text{g}$ 、不純物成分が $1.0 \sim 5\%$ の炭化ケイ素粉末を出発原料を所望の形状に成形、乾燥後、 $1600 \sim 2200^\circ\text{C}$ の温度範囲で焼成することを特徴とするものである。

【0012】即ち、従来、比表面積が $5 \text{ m}^2/\text{g}$ 以上と大きく、不純物が少なく、又は／及び、平均粒径が $1 \mu\text{m}$ 以下と小さい炭化ケイ素($\beta\text{-SiC}$)粉末を出発原料とすることによってのみ触媒担体用として優れた多孔質炭化ケイ素焼結体が製造出来るものと思われて来た。即ち、平均粒径が $1 \mu\text{m}$ 以下の超微粉が焼結(ネッ

キング)して成長をすることにより、成形体の中で炭化ケイ素粉末が占有している体積が減少し、これにより細孔容積が増大すると同時に触媒担体又は／およびフィルターとして最適な連続気孔が出来ること、そして、原料粉末に適した最高焼成温度で焼成することにより、最適な気孔径の触媒担体が製造出来るものであって、不純物が少しでも多かたり、比表面積又は／および平均粒径が少しでも大きいと不適當であるとされて来た。

【0013】しかし、多孔質炭化ケイ素焼結体の製造について、各種生成条件と得られた生成粒との関係を詳細に研究した結果、比表面積が小さく、不純物が一定範囲で大きく、且つ、好ましくは不純物として、焼結助剤として作用するホウ素化合物が金属換算で $0.1 \sim 0.5\%$ 範囲で混入して炭化ケイ素($\alpha\text{-SiC}$)粉末を用いることにより触媒担体として優れた性能を有する多孔質炭化ケイ素質焼結体からなる触媒担体を経済的に製造することを見出したのである。すなわち、本発明においては、比表面積が $0.1 \sim 5 \text{ m}^2/\text{g}$ 、不純物成分が $1.0 \sim 5\%$ の炭化ケイ素粉末を使用するのであって、この比表面積の範囲において、比表面積が大きくなるにしたがって、その不純物成分が $1.0 \sim 5\%$ の範囲で少なく、且つ、最高焼成温度範囲の $1600 \sim 2200^\circ\text{C}$ において、高くすることが重要である。

【0014】比表面積が $0.1 \text{ m}^2/\text{g}$ より小さくなると、触媒担体の細孔容積および比表面積が小さくなり過ぎるので好ましくない。又、比表面積が $5 \text{ m}^2/\text{g}$ 以上の炭化ケイ素粉末を出発原料とすることは出発原料コストが著しく高くなり、好ましくない。不純物成分が 1% 以下の出発原料とすると、前記同様に経済的でない。また、 5% 以上とすると触媒担体の高温強度が低下する等の欠点が生じて好ましくない。このような出発原料は、古くから製造されている $\alpha\text{-SiC}$ の製造方法(アチソン法)によって容易に得ることができる。

【0015】焼成温度について、最高焼成温度を 1600°C 以下にすると前記同様に触媒担体の高温強度が低下する等の欠点が生じて好ましくなく、 2200°C 以上では製造コストが著しく高くなってしまふ。

【0016】尚、不純物成分中にはホウ素化合物が金属換算で $0.1 \sim 0.5\%$ 含有していることが好ましい。すなわち、ホウ素化合物は、前記炭化ケイ素粉出発原料の焼結助剤としての作用をするものである。 0.1% 以下では助剤としての効果が小さ過ぎ、 0.5% 以上になると成形体の収縮率が大きくなってしまふので焼成最高温度を低下させなければならなくなるが、焼成最高温度を低下させることは、触媒担体の高温強度の劣化等の欠点を生ずることとなる。次に、実施例をもって、更に本発明を具体的に説明する。

【0017】

【実施例】

50 実施例 1

比表面積 $0.7 \text{ m}^2/\text{g}$ で不純物含有量 3.0% (B、Cが 0.3%)の α -炭化ケイ素を 70% と、比表面積 $12 \text{ m}^2/\text{g}$ で不純物含有量 1.0% の β -炭化ケイ素を 30% とを混合した炭化ケイ素粉末 100 重量部に、水 22 重量部、メチルセルロース 重量部、グリセリン 2 部、界面活性剤 4 部とを混合、混練した坯土を押出成形機で外径 140 mm 、長さ 70 mm 、セル厚 0.43 mm 、セル数 170 コ/ in^2 の成形体を成形・乾燥後、 1800°C で 4 時間焼成した結果、気孔率 45% 、焼成収縮率 2% 、平均気孔径 $15 \mu\text{m}$ 、曲げ強度 4

$50 \text{ kg}/\text{cm}^2$ (セル温度) のハニカム構造の多孔質炭化ケイ素質触媒担体が得られた。

【0018】

【発明の効果】以上述べたように、本発明においては、比表面積が $0.1 \sim 5 \text{ m}^2/\text{gr}$ 、不純物成分が $1.0 \sim 5\%$ の炭化ケイ素粉末を出発原料とし、これを $1600^\circ\text{C} \sim 2200^\circ\text{C}$ の温度範囲で焼成することによって、従来の多孔質炭化ケイ素よりなる触媒担体に比して高い強度、大きな比表面積と細孔容積を有する等の特性を有する触媒担体が得られる。

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DETAILED DESCRIPTION

[Detailed Description of the Invention]

[0001]

[Industrial Application] This invention relates to the manufacture approach of catalyst support which consists of a porosity silicon carbide sintered compact which has high reinforcement, and a big specific surface area and pore volume.

[0002]

[Description of the Prior Art] Silicon carbide is an usable ingredient large as corrosion-resistant ingredients, such as pump components of the solution which has the property which was physically [chemically / an elevated temperature, high reinforcement, etc. / and] excellent in a high degree of hardness, the outstanding abrasion resistance, the outstanding oxidation resistance, the outstanding corrosion resistance, good thermal conductivity, a low coefficient of thermal expansion, high thermal shock resistance, and a list, and has corrosive, such as thermal-protection-system ingredients, such as refractory material abrasion resistance materials, such as mechanical seal and a bearing, and for elevated-temperature furnaces, and a heat exchanger or the combustion tube,, an acid, and alkali, / strong].

[0003] The nature sintered compact of porosity silicon carbide which consists of silicon carbide which has these properties and pore which has the permeability which the crystal forms, i.e., an open pore, on the other hand The heatproof [the description of said silicon carbide is employed efficiently and] under an elevated-temperature ambient atmosphere, an oxidizing atmosphere, and/or a corrosive ambient atmosphere, It is available as a corrosion-resistant matter separation ingredient, for example, can use as a filter used for removal of particle matter, such as particle carbon contained in elevated-temperature gases, such as an internal combustion engine's exhaust gas, especially exhaust gas of a diesel power plant. Furthermore, an inflammable carbon particle is made to burn, when this porosity silicon carbide filter front face is made to support the catalyst component for oxidation reaction, it will be possible on it to also make gas convert, and this porosity silicon carbide filter will function on it also as a heatproof and *****-proof in this case.

[0004] Moreover, from a viewpoint of an environmental pollution control, in the field of industrial use burners, such as an internal combustion engine and a boiler for gas turbines, researches and developments of a low-NOx-combustion technique are done, and the catalyzed combustion technique using a combustion catalyst as one mainly attracts attention recently. The most important element in development of this catalyzed combustion technique is development of a catalyst, and development of the support for ranking with development of the activity oxidation anti-application catalyst matter in development of a catalyst, and distributing and supporting an active ingredient is very important for it.

[0005] For surface area to be large in order to make the combustion reaction which advances on a catalyst front face, i.e., oxidation reaction, occur quickly in the support for combustion catalysts In addition, the thing for which it has the good thermal conductivity which can transmit and remove heat of reaction to generate effectively, And in order to make the mass transfer in catalyst pore perform effectively, passage resistance of fluids, such as gas, is small, Namely, the thing for which it has

mechanical strength with sufficient that it is strong to that pore volume is large and the abrasion by the collision further between Plastic solids, i.e., wear, and Plastic solid itself, And things for which many demands are satisfied -- these properties are stable to use of long duration -- are required. Such requirements are being able to say in [support / which is used at an elevated temperature / the catalyst support for chemical reactions not only a combustion catalyst but accompanied by generating of heat of reaction generally, or / for catalysts] common.

[0006] On the other hand, as the manufacture approach of the nature sintered compact of porosity silicon carbide (1) After adding and fabricating binding material, such as glassiness flux or argillaceous, to the silicon carbide particle used as the aggregate, How to burn, harden and manufacture the Plastic solid at the temperature which said binding material fuses, (2) After mixing and fabricating the silicon carbide particle of a big and rough grain, and a detailed silicon carbide particle, While adding a carbonaceous binder, without adding or adding carbon powder to the approach of calcinating and manufacturing at an elevated temperature 2000 degrees C or more, or the silicon carbide powder currently indicated by invention of (3) JP,48-39515,A The free carbon from the binder generated at the time of carbon powder and baking and the clay genius powder of the amount of theory which reacts are added and formed. The manufacture approach of the homogeneity porosity recrystallization silicon carbide object characterized by heating at 1900-2400 degrees C in the carbon powder in this Plastic solid, and silicon-izing the carbon content in a Plastic solid after an appropriate time and (4) specific surface area above 3m²/g The sum total of the content of boron, aluminum, and iron converts into an element, and the approach of calcinating and manufacturing at 1600-2200 degrees C etc. is learned conventionally after fabricating the silicon carbide powder which is 0.3 or less % of the weight in a desired configuration.

[0007]

[Problem(s) to be Solved by the Invention] However, there is the following problem in the conventional technique mentioned above.

(1) since binding material fuse the approach of burn, harden and manufacture that Plastic solid at the temperature which said binding material fuse, at low temperature (before or after 1000-1400 degrees C) after add and fabricate binding material, such as glassiness flux or argillaceous, to the silicon carbide particle used as the aggregate, a porous body have the fault that the use in the field as which reinforcement not only fall remarkably, but it change this temperature region, especially near glass transition temperature, and chemical resistance and oxidation resistance be require be restrict.

[0008] (2) How to calcinate and manufacture at an elevated temperature 2000 degrees C or more, after mixing and fabricating the silicon carbide particle of a big and rough grain, and a detailed silicon carbide particle, Or while adding a carbonaceous binder, without adding or adding carbon powder to the silicon carbide powder currently indicated by invention of (3) JP,48-39515,A The free carbon from the binder generated at the time of this carbon powder and baking and the clay genius powder of the amount of theory which reacts are added and formed. The manufacture approach of the homogeneity porosity recrystallization silicon carbide object characterized by heating at 1900-2400 degrees C in the carbon powder in this Plastic solid, and silicon-izing the carbon content in a Plastic solid after an appropriate time The porosity silicon carbide aggregate and its aggregate are covered, and the nature binding material of silicon carbide or the carbonaceous binding material, and the gap which combine an aggregate comrade are constituted. Most gaps of said porosity, i.e., an open pore, need many aggregate particles and becoming big and rough particles at the time of shaping, and, as a result, its point of contact of an aggregate particle decreases, porous reinforcement falls remarkably and, moreover, specific surface area becomes a remarkable small thing below by 0.5m²/g.

[0009] On the other hand, in order to consider as a porous body with high reinforcement, it is required to mix with coarse grain, middle whenever/, or a particle moderately, and to form the particle size blending of the aggregate, consequently when it is remarkable, small and extreme at most at 0.1 ml/g, some open pores tend to blockade the pore volume of a porous body. For this reason, such resistance at the time of a fluid passing a porous body becomes it is remarkable and disadvantageous, when becoming remarkably high and using as the filter for matter separation, and ***** . Therefore, it has the nature sintered compact of silicon carbide, i.e., reinforcement with easy handling, which has a property

suitable as catalyst support, and moreover, pore volume is larger than $0.2\text{m}^2/\text{g}$, and the nature sintered compact of porosity silicon carbide with a bigger specific surface area than $3\text{m}^2/\text{g}$ does not exist.

[0010] (4) Although an applicant has the approach proposed previously as an approach of manufacturing the porous body which has the above properties The approach which specific surface area is more than $3\text{m}^2/\text{g}$ for this approach, and the content of boron, aluminum, and iron calcinates at 1400-2000 degrees C using 0.3 or less % of the weight of a thing in total is a high grade. and it not only becomes a cost rise remarkably [since superfines silicon carbide is used as the start raw material], but The fault which a desiccation crack tends to generate by the rate of drying shrinkage of a Plastic solid becoming large is carried out.: Only the thing of the pore diameter not more than several micrometer order or it is obtained to a pore diameter being suitable for 15-micrometer order like the perch curate filter in diesel-power-plant exhaust gas. Although what is necessary is just to have performed burning temperature above 2000 degrees C as an approach of enlarging a pore diameter, by said approach, it had a fault, like degradation on the strength will be caused with abnormality grain growth etc.

[0011]

[Means for Solving the Problem] This invention cancels said conventional fault, and improves, and in order to supply the catalyst support which consists of a nature sintered compact of porosity silicon carbide which has a property required as heat-resistant catalyst support, specific surface area is characterize by calcinate a start raw material after shaping and desiccation in a 1600 degrees C - 2200 degrees C temperature requirement in the configuration of a request of 1.0 - 5% of silicon carbide powder of $0.1\text{-}5\text{m}^2/\text{gr}$ and an impurity component.

[0012] That is, conventionally, specific surface area is as large as more than $5\text{m}^3/\text{gr}$, and it has been thought that the porosity silicon carbide sintered compact in which mean particle diameter was [that there are few impurities] excellent as an object for catalyst support only by using 1 micrometer or less and small silicon carbide (beta-SiC) powder as a start raw material can be manufactured. Namely, when mean particle diameter grows by superfines 1 micrometer or less sintering (necking) Catalyst support or/and continuation pore optimal as a filter are made at the same time the volume which silicon carbide powder occupies in a Plastic solid decreases and pore volume increases by this, And by calcinating with the highest burning temperature suitable for raw material powder, the catalyst support of the optimal pore diameter can be manufactured, there are more impurities, or if specific surface area or/and mean particle diameter are larger, it will have been supposed that it is unsuitable.

[0013] As a result of studying the relation between various generation conditions and the obtained generation grain in a detail about manufacture of a porosity silicon carbide sintered compact, specific surface area is small. An impurity preferably greatly in the fixed range however, as an impurity It found out manufacturing economically the catalyst support which consists of a nature sintered compact of porosity silicon carbide which has the engine performance which was excellent as catalyst support by the boron compound which acts as sintering acid mixing in the range 0.1 to 0.5% by metal conversion, and using silicon carbide (alpha-SiC) powder. That is, in this invention, there are few those impurity components in 1.0 - 5% of range, and it is important in 1600-2200 degrees C of the highest burning-temperature range to make it high as $0.1\text{-}5\text{m}^2/\text{gr}$ and an impurity component use [specific surface area] 1.0 - 5% of silicon carbide powder and specific surface area becomes large in the range of this specific surface area.

[0014] If specific surface area becomes smaller than $0.1\text{m}^2/\text{gr}$, since the pore volume and specific surface area of catalyst support will become small too much, it is not desirable. Moreover, start raw material cost becomes remarkably high, and it does not have it that specific surface area uses the silicon carbide powder more than $5\text{m}^2/\text{gr}$ as a start raw material. [desirable] If an impurity component considers as 1% or less of start raw material, it is not economical to said this appearance. Moreover, if 5% or more, the fault of the high temperature strength of catalyst support falling generates and is not desirable. Such a start raw material can be easily obtained by the manufacture approach (Acheson process) of alpha-SiC manufactured for many years.

[0015] About burning temperature, if the highest burning temperature is made into 1600 degrees C or less, the fault of the high temperature strength of catalyst support falling to said this appearance will

arise, and a manufacturing cost will become remarkably high above 2200 degrees C preferably.

[0016] In addition, it is desirable that the boron compound contains 0.1 to 0.5% by metal conversion in an impurity component. That is, a boron compound carries out the operation as sintering acid of said silicon carbide powder start raw material. Although it must stop having to reduce a baking maximum temperature at 0.1% or less since contraction of a Plastic solid will become large if the effectiveness as an assistant is too small and makes it to 0.5% or more, reducing a baking maximum temperature will produce faults, such as degradation of the high temperature strength of catalyst support. Next, it has an example and this invention is explained further concretely.

[0017]

[Example]

Example alpha-silicon carbide of 3.0% of impurity contents (B₄C is 0.3%) by 1 specific-surface-area of 0.7m²/g 70%, In the silicon carbide powder 100 weight section which mixed 30%, beta-silicon carbide of 1.0% of impurity contents by specific-surface-area of 12m²/g The water 22 weight section, methyl cellulose The weight section, the glycerol 2 section, The plastic matter which mixed the surfactant 4 section and was kneaded with an extruding press machine Outer-diameter 140 m/m, The Plastic solid of 2 die-length 70 m/m, cel thickness 0.43 m/m, and 170 cel numbers/inch After shaping / desiccation, As a result of calcinating at 1800 degrees C for 4 hours, the nature catalyst support of porosity silicon carbide of honeycomb structure with 45% of porosity, the burning shrinkage of 2%, an average pore diameter [of 15 micrometers], and a flexural strength of 450kg/cm² (cel temperature) was obtained.

[0018]

[Effect of the Invention] As stated above, in this invention, the catalyst support in which specific surface area has properties, such as having high reinforcement, and a big specific surface area and pore volume as compared with the catalyst support which consists of conventional porosity silicon carbide by 0.1-5m²/gr and an impurity component using 1.0 - 5% of silicon carbide powder as a start raw material, and calcinating this in a 1600 degrees C - 2200 degrees C temperature requirement, is obtained.

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CLAIMS

[Claim(s)]

[Claim 1] The manufacture approach of catalyst support that specific surface area is characterized by for 0.1-5m²/gr and an impurity component using 1.0 - 5% of silicon carbide powder as a start raw material, and calcinating after shaping and desiccation in the configuration of a request of this in a 1600 degrees C - 2200 degrees C temperature requirement.

[Claim 2] The manufacture approach of the catalyst support given in the 1st term of a claim characterized by the boron compound containing 0.1 to 0.5% by metal conversion in an impurity component.

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TECHNICAL FIELD

[Industrial Application] This invention relates to the manufacture approach of catalyst support which consists of a porosity silicon carbide sintered compact which has high reinforcement, and a big specific surface area and pore volume.

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PRIOR ART

[Description of the Prior Art] Silicon carbide is an usable ingredient large as corrosion-resistant ingredients, such as pump components of the solution which has the property which was physically [chemically / an elevated temperature, high reinforcement, etc. / and] excellent in a high degree of hardness, the outstanding abrasion resistance, the outstanding oxidation resistance, the outstanding corrosion resistance, good thermal conductivity, a low coefficient of thermal expansion, high thermal shock resistance, and a list, and has corrosive, such as thermal-protection-system ingredients, such as refractory material abrasion resistance materials, such as mechanical seal and a bearing, and for elevated-temperature furnaces, and a heat exchanger or the combustion tube,, an acid, and alkali, / strong].

[0003] The nature sintered compact of porosity silicon carbide which consists of silicon carbide which has these properties and pore which has the permeability which the crystal forms, i.e., an open pore, on the other hand The heatproof [the description of said silicon carbide is employed efficiently and] under an elevated-temperature ambient atmosphere, an oxidizing atmosphere, and/or a corrosive ambient atmosphere, It is available as a corrosion-resistant matter separation ingredient, for example, can use as a filter used for removal of particle matter, such as particle carbon contained in elevated-temperature gases, such as an internal combustion engine's exhaust gas, especially exhaust gas of a diesel power plant. Furthermore, an inflammable carbon particle is made to burn, when this porosity silicon carbide filter front face is made to support the catalyst component for oxidation reaction, it will be possible on it to also make gas convert, and this porosity silicon carbide filter will function on it also as a heatproof and *****-proof in this case.

[0004] Moreover, from a viewpoint of an environmental pollution control, in the field of industrial use burners, such as an internal combustion engine and a boiler for gas turbines, researches and developments of a low-NOx-combustion technique are done, and the catalyzed combustion technique using a combustion catalyst as one mainly attracts attention recently. The most important element in development of this catalyzed combustion technique is development of a catalyst, and development of the support for ranking with development of the activity oxidation anti-application catalyst matter in development of a catalyst, and distributing and supporting an active ingredient is very important for it.

[0005] For surface area to be large in order to make the combustion reaction which advances on a catalyst front face, i.e., oxidation reaction, occur quickly in the support for combustion catalysts In addition, the thing for which it has the good thermal conductivity which can transmit and remove heat of reaction to generate effectively, And in order to make the mass transfer in catalyst pore perform effectively, passage resistance of fluids, such as gas, is small, Namely, the thing for which it has mechanical strength with sufficient that it is strong to that pore volume is large and the abrasion by the collision further between Plastic solids, i.e., wear, and Plastic solid itself, And things for which many demands are satisfied -- these properties are stable to use of long duration -- are required. Such requirements are being able to say in [support / which is used at an elevated temperature / the catalyst support for chemical reactions not only a combustion catalyst but accompanied by generating of heat of reaction generally, or / for catalysts] common.

[0006] On the other hand, as the manufacture approach of the nature sintered compact of porosity silicon carbide (1) After adding and fabricating binding material, such as glassiness flux or argillaceous, to the silicon carbide particle used as the aggregate, How to burn, harden and manufacture the Plastic solid at the temperature which said binding material fuses, (2) After mixing and fabricating the silicon carbide particle of a big and rough grain, and a detailed silicon carbide particle, While adding a carbonaceous binder, without adding or adding carbon powder to the approach of calcinating and manufacturing at an elevated temperature 2000 degrees C or more, or the silicon carbide powder currently indicated by invention of (3) JP,48-39515,A The free carbon from the binder generated at the time of carbon powder and baking and the cay genius powder of the amount of theory which reacts are added and formed. The manufacture approach of the homogeneity porosity recrystallization silicon carbide object characterized by heating at 1900-2400 degrees C in the carbon powder in this Plastic solid, and silicon-izing the carbon content in a Plastic solid after an appropriate time and (4) specific surface area above 3m²/g The sum total of the content of boron, aluminum, and iron converts into an element, and the approach of calcinating and manufacturing at 1600-2200 degrees C etc. is learned conventionally after fabricating the silicon carbide powder which is 0.3 or less % of the weight in a desired configuration.

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EFFECT OF THE INVENTION

[Effect of the Invention] As stated above, in this invention, the catalyst support in which specific surface area has properties, such as having high reinforcement, and a big specific surface area and pore volume as compared with the catalyst support which consists of conventional porosity silicon carbide by 0.1-5m²/gr and an impurity component using 1.0 - 5% of silicon carbide powder as a start raw material, and calcinating this in a 1600 degrees C - 2200 degrees C temperature requirement, is obtained.

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TECHNICAL PROBLEM

[Problem(s) to be Solved by the Invention] However, there is the following problem in the conventional technique mentioned above.

(1) since binding material fuse the approach of burn, harden and manufacture that Plastic solid at the temperature which said binding material fuse, at low temperature (before or after 1000-1400 degrees C) after add and fabricate binding material, such as glassiness flux or argillaceous, to the silicon carbide particle used as the aggregate, a porous body have the fault that the use in the field as which reinforcement not only fall remarkably, but it change this temperature region, especially near glass transition temperature, and chemical resistance and oxidation resistance be require be restrict.

[0008] (2) How to calcinate and manufacture at an elevated temperature 2000 degrees C or more, after mixing and fabricating the silicon carbide particle of a big and rough grain, and a detailed silicon carbide particle, Or while adding a carbonaceous binder, without adding or adding carbon powder to the silicon carbide powder currently indicated by invention of (3) JP,48-39515,A The free carbon from the binder generated at the time of this carbon powder and baking and the cay genius powder of the amount of theory which reacts are added and formed. The manufacture approach of the homogeneity porosity recrystallization silicon carbide object characterized by heating at 1900-2400 degrees C in the carbon powder in this Plastic solid, and silicon-izing the carbon content in a Plastic solid after an appropriate time The porosity silicon carbide aggregate and its aggregate are covered, and the nature binding material of silicon carbide or the carbonaceous binding material, and the gap which combine an aggregate comrade are constituted. Most gaps of said porosity, i.e., an open pore, need many aggregate particles and becoming big and rough particles at the time of shaping, and, as a result, its point of contact of an aggregate particle decreases, porous reinforcement falls remarkably and, moreover, specific surface area becomes a remarkable small thing below by 0.5m²/g.

[0009] On the other hand, in order to consider as a porous body with high reinforcement, it is required to mix with coarse grain, middle whenever/, or a particle moderately, and to form the particle size blending of the aggregate, consequently when it is remarkable, small and extreme at most at 0.1 ml/g, some open pores tend to blockade the pore volume of a porous body. For this reason, such resistance at the time of a fluid passing a porous body becomes it is remarkable and disadvantageous, when becoming remarkably high and using as the filter for matter separation, and ***** Therefore, it has the nature sintered compact of silicon carbide, i.e., reinforcement with easy handling, which has a property suitable as catalyst support, and moreover, pore volume is larger than 0.2m²/g, and the nature sintered compact of porosity silicon carbide with a bigger specific surface area than 3m²/g does not exist.

[0010] (4) Although an applicant has the approach proposed previously as an approach of manufacturing the porous body which has the above properties The approach which specific surface area is more than 3m²/g for this approach, and the content of boron, aluminum, and iron calcinates at 1400-2000 degrees C using 0.3 or less % of the weight of a thing in total is a high grade. and it not only becomes a cost rise remarkably [since superfines silicon carbide is used as the start raw material], but The fault which a desiccation crack tends to generate by the rate of drying shrinkage of a Plastic solid becoming large is carried out.: Only the thing of the pore diameter not more than several micrometer order or it is obtained

to a pore diameter being suitable for 15-micrometer order like the percurate filter in diesel-power-plant exhaust gas. Although what is necessary is just to have performed burning temperature above 2000 degrees C as an approach of enlarging a pore diameter, by said approach, it had a fault, like degradation on the strength will be caused with abnormality grain growth etc.

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MEANS

[Means for Solving the Problem] This invention cancels said conventional fault, and improves, and in order to supply the catalyst support which consists of a nature sintered compact of porosity silicon carbide which has a property required as heat-resistant catalyst support, specific surface area is characterized by calcinate a start raw material after shaping and desiccation in a 1600 degrees C - 2200 degrees C temperature requirement in the configuration of a request of 1.0 - 5% of silicon carbide powder of 0.1-5m²/gr and an impurity component.

[0012] That is, conventionally, specific surface area is as large as more than 5m³/gr, and it has been thought that the porosity silicon carbide sintered compact in which mean particle diameter was [that there are few impurities] excellent as an object for catalyst support only by using 1 micrometer or less and small silicon carbide (beta-SiC) powder as a start raw material can be manufactured. Namely, when mean particle diameter grows by superfines 1 micrometer or less sintering (necking) Catalyst support or/and continuation pore optimal as a filter are made at the same time the volume which silicon carbide powder occupies in a Plastic solid decreases and pore volume increases by this, And by calcinating with the highest burning temperature suitable for raw material powder, the catalyst support of the optimal pore diameter can be manufactured, there are more impurities, or if specific surface area or/and mean particle diameter are larger, it will have been supposed that it is unsuitable.

[0013] As a result of studying the relation between various generation conditions and the obtained generation grain in a detail about manufacture of a porosity silicon carbide sintered compact, specific surface area is small. An impurity preferably greatly in the fixed range however, as an impurity It found out manufacturing economically the catalyst support which consists of a nature sintered compact of porosity silicon carbide which has the engine performance which was excellent as catalyst support by the boron compound which acts as sintering acid mixing in the range 0.1 to 0.5% by metal conversion, and using silicon carbide (alpha-SiC) powder. That is, in this invention, there are few those impurity components in 1.0 - 5% of range, and it is important in 1600-2200 degrees C of the highest burning-temperature range to make it high as 0.1-5m²/gr and an impurity component use [specific surface area] 1.0 - 5% of silicon carbide powder and specific surface area becomes large in the range of this specific surface area.

[0014] If specific surface area becomes smaller than 0.1m²/gr, since the pore volume and specific surface area of catalyst support will become small too much, it is not desirable. Moreover, start raw material cost becomes remarkably high, and it does not have it that specific surface area uses the silicon carbide powder more than 5m²/gr as a start raw material. [desirable] If an impurity component considers as 1% or less of start raw material, it is not economical to said this appearance. Moreover, if 5% or more, the fault of the high temperature strength of catalyst support falling generates and is not desirable. Such a start raw material can be easily obtained by the manufacture approach (Acheson process) of alpha-SiC manufactured for many years.

[0015] About burning temperature, if the highest burning temperature is made into 1600 degrees C or less, the fault of the high temperature strength of catalyst support falling to said this appearance will arise, and a manufacturing cost will become remarkably high above 2200 degrees C preferably.

[0016] In addition, it is desirable that the boron compound contains 0.1 to 0.5% by metal conversion in an impurity component. That is, a boron compound carries out the operation as sintering aid of said silicon carbide powder start raw material. Although it must stop having to reduce a baking maximum temperature at 0.1% or less since contraction of a Plastic solid will become large if the effectiveness as an assistant is too small and makes it to 0.5% or more, reducing a baking maximum temperature will produce faults, such as degradation of the high temperature strength of catalyst support. Next, it has an example and this invention is explained further concretely.

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EXAMPLE

[Example]

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